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HIGH-TEMPERATURE CYCLIC FATIGUE-CRACK GROWTH IN MONOLITHIC Ti_3SiC_2 CERAMICS

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Abstract

The cyclic fatigue behavior of reactive hot-pressed Ti_3SiC_2 ceramics are examined at temperatures from ambient to 1200°C with the objective of characterizing the high-temperature mechanisms controlling crack growth. Comparisons are made of two monolithic Ti_3SiC_2 materials with fine- (3-10 µm) and coarse-grained (70-300 µm) microstructures. Results indicate that the ΔK_{th} fatigue thresholds are not substantially changed between 25° and 1100°C; however, there is a sharp decrease in ΔK_{th} at 1200°C (above the "ductile-brittle" transition temperature), where significant high-temperature deformation and damage are first apparent. Of the two microstructures, the coarse-grained Ti_3SiC_2 exhibits substantially higher cyclic-crack growth resistance at both ambient and elevated temperatures. This results from an enhanced effect in the coarser grained microstructure of crack bridging in the crack wake from both grains and lamellae within grains, and from the correspondingly more tortuous crack path.

Introduction

The polycrystalline ternary carbide, Ti_3SiC_2, exhibits a surprising combination of properties for a ceramic; for example, it displays high toughness (K_c > 8 MPa√m) and a ratio of hardness (~4 GPa) to elastic modulus (~320 GPa) more typical of a ductile material. Processed by reactive hot-pressing techniques from powders of Ti, SiC and graphite, Ti_3SiC_2 also shows a range of inelastic deformation modes not typically seen in ceramics at room temperature (1-6), including grain bending, grain buckling, and significant amounts of basal slip. Indeed, in general Ti_3SiC_2 appears to be one of the most damage tolerant of all non-transforming monolithic ceramics (1-7).

In Ti_3SiC_2, basal plane dislocations are mobile and multiply at room temperature. They are confined to two orthogonal directions: basal plane arrays (wherein the dislocations exist on identical slip planes) and walls or kink boundaries. Thus, in addition to regular slip, mechanisms for ambient temperature plastic deformation in Ti_3SiC_2 are thought to involve the readjustment of local stress and strain fields from kink band (boundaries) formation, buckling and delamination of *individual* grains, the delamination and associated damage being contained by the kink boundaries (6). The delaminations typically occur at the intersection of the walls and arrays and result in the annihilation of the latter (6). It is this containment of damage that is believed to be the major source of damage tolerance in Ti_3SiC_2; in fact, the plastic behavior in general is unusual for carbides and is believed to be due to its layered structure and the metallic nature of the bonding (6).

With such intrinsic deformation and toughness properties, Ti_3SiC_2 clearly offers some potential for many structural applications; however, if this is to be realized, it is important that some assessment be made of its crack-growth behavior, particularly at elevated temperatures. It is the objective of the present paper to examine the subcritical crack-growth characteristics of Ti_3SiC_2 under cyclic loads, in both fine- and coarse-grained microstructural conditions and at temperatures from ambient to 1200°C, with the objective of defining the salient damage and crack-tip shielding mechanisms that govern crack advance.

Background

The fracture toughness and cyclic fatigue-crack growth behavior of monolithic Ti_3SiC_2 at ambient temperatures was first characterized by Gilbert *et al.* (7) in both fine- (3-10 µm) and coarse-grained (50-200 µm) conditions. Fatigue-crack growth thresholds, ΔK_{th}, were found to be as high as 6 and 9 MPa√m, respectively, in the fine and coarse-grained structures; corresponding fracture toughnesses, K_c were measured (at the peak of the R-curve) at, respectively, 9.5 and 16 MPa√m. The high toughness (the K_c value for the coarse-grained structure is thought to be the highest ever reported for a monolithic, non-transforming ceramic) was found to be associated with a profusion of crack-bridging processes active in the crack wake. Indeed, Ti_3SiC_2 displayed a far larger degree of grain bridging and sliding (3,4,7) than has been observed in other ceramics such as Al_2O_3, Si_3N_4, and SiC (e.g., 8-10), possibly because the deformation processes observed in individual grains enhanced grain bridging by increasing pullout distances and suppressing grain rupture. Fatigue-crack growth, on the other hand, was associated with the cyclic loading induced degradation of such bridging; substantial evidence was found for wear degradation at active bridging sites behind the crack tip, particularly in the coarser-grained structure. Although significant progress has been made in understanding the mechanical behavior of polycrystalline Ti_3SiC_2 at ambient temperatures, far less is understood about its properties at elevated temperatures. Its mechanical response in tension up to 1300°C has been documented (11); however, nothing is known about its fatigue-crack growth behavior at these temperatures.

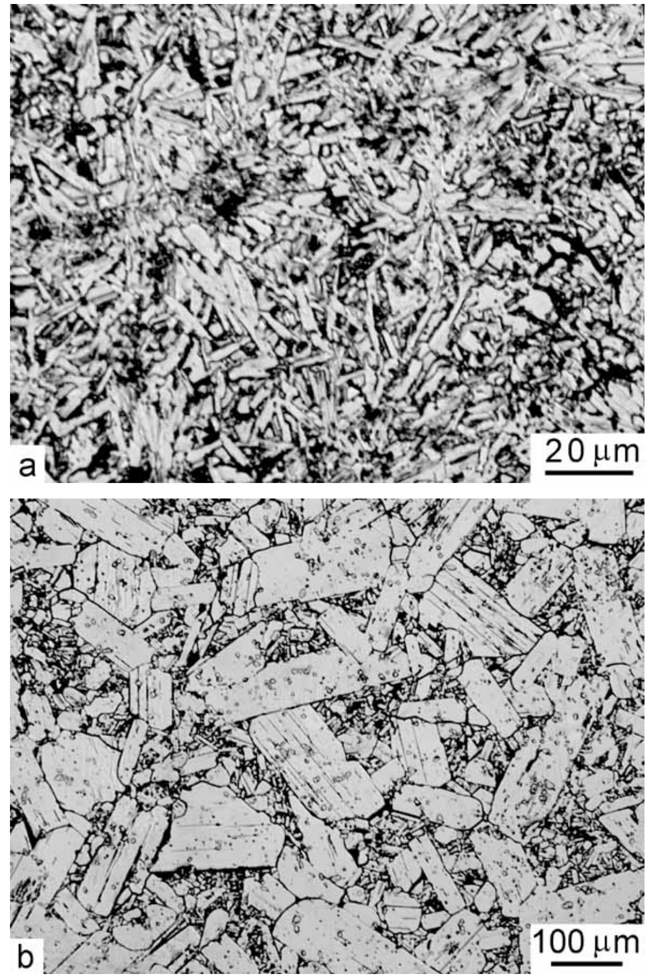

Figure 1: Optical micrographs of the microstructures in Ti_3SiC_2 with (a) fine grains (3-10 μm) and (b) coarse grains (70-300 μm) (etched in a 1:1:1 by volume $HF:HNO_3:H_2O$ solution).

Experimental Procedures

Material processing

Ti_3SiC_2 samples were fabricated by a reactive hot isostatic pressing technique using TiH_2 (-325 mesh, 99.99%), SiC (grain size ~ 20 μm, 99.5%) and graphite (grain size ~ 1 μm, 99%) starting powders. Powders with the desired stoichiometry were mixed in a ball mill, cold-isostatically pressed at 200 MPa, and annealed at 900°C for 4 hr *in vacuo* to remove hydrogen. Samples were then sealed in glass under vacuum and hot-isostatically pressed for 4 hr at 1400°C and 1600°C to form the fine- and coarse-grained microstructures, respectively. The coarse-grained structure consisted of large plate-like grains of diameter 70-300 μm and thickness 5-30 μm, whereas grains in the fine-grained microstructure had a diameter of ~7 μm and were ~3 μm thick (Fig. 1). Each individual grain consists of thin layers, with a width of 2 –3 μm, normal to the basal planes. Further details on the procedures for fabricating Ti_3SiC_2 and its microstructural evolution have been reported elsewhere (12,13).

Cyclic fatigue testing

Cyclic fatigue-crack growth tests were performed using 2.9-mm thick compact-tension C(T) specimens (of width ~19 mm); this geometry conforms to ASTM Standard E-647 for fatigue-crack growth rate measurements. Specimens were cycled at a constant load ratio (ratio of minimum to maximum applied loads) of R = 0.1 and a loading frequency of ν = 25 Hz (sinusoidal), using automated stress-intensity K control. Growth rates were monitored under decreasing ΔK conditions with a normalized K-gradient ($1/K \cdot dK/da$) set to –0.08 mm^{-1}. Prior to testing, the C(T) samples were polished (with ~1 μm diamond paste) and fatigue pre-cracked at room temperature for several millimeters beyond the half-chevron-shaped, starter notch. The latter notch geometry is used to facilitate crack initiation in brittle materials.

At elevated temperatures (1100º and 1200ºC), testing was performed on a computer-controlled servo-hydraulic mechanical testing system in flowing gaseous argon at atmospheric pressure in an environmental chamber/furnace, heated by graphite elements that maintain temperature to within ±1°C. Heating and cooling rates were kept at 10°C/min to minimize any thermal shock effects. After reaching the desired temperature and prior to commencing the test, the furnace temperature was kept constant for 1-3 hr to permit the thermal equilibrium of the system.

At ambient temperatures, fatigue-crack growth rates were continuously measured using the standard back-face strain elastic-unloading compliance method, using a 350 Ω strain gauge affixed to the back surface of the specimen. This method could not be readily used at elevated temperatures; however, as Ti_3SiC_2 has good electrical conductivity at all temperatures, crack lengths at 1100° and 1200°C could be monitored *in situ* using the electrical-potential method. Full details of the use of electrical-potential methods for crack length monitoring in ceramics are given in refs. 14,15. To verify both elevated- and room-temperature measurements, readings were checked periodically using an optical microscope. Cyclic fatigue-crack growth data are presented in terms of the growth rate per cycle, da/dN, as a function of the applied stress-intensity range, ΔK (= K_{max} - K_{min}), the latter being computed using standard linear-elastic handbook solutions.

Crack profiles of selected fatigue samples, taken at mid-section of the test piece perpendicular to the fracture surface, were examined in a scanning electron microscope (SEM).

Results and Discussion

The variation in fatigue-crack growth rates, da/dN, with applied stress-intensity range, ΔK, in Ti_3SiC_2 at temperatures of 25°, 1100° and 1200°C (R = 0.1, ν = 25 Hz - sine wave), is shown in Fig. 2 for the fine- and coarse-grained microstructures.

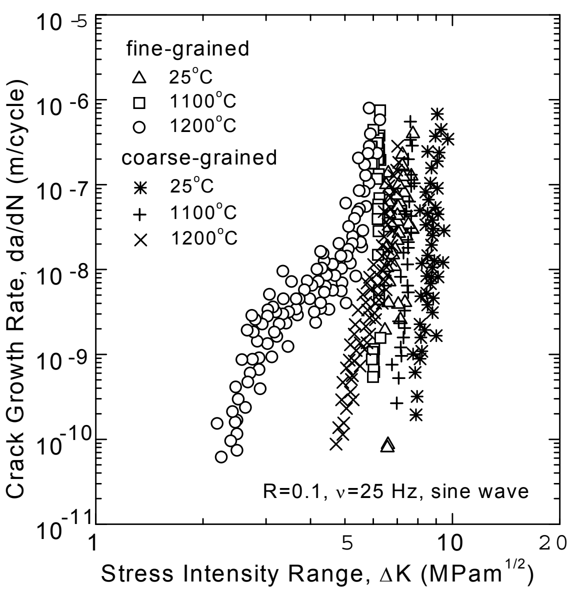

Figure 2: Cyclic fatigue-crack growth rates, da/dN, at R = 0.1 and ν = 25 Hz (sine wave) in Ti_3SiC_2 with fine- and coarse-grained microstructures, as a function of the applied stress-intensity range ΔK at temperatures of 25°, 1100° and 1200°C.

Characteristic of ceramic materials at low homologous temperatures (16,17), growth rates in both Ti_3SiC_2 microstructures at 25°C display a marked sensitivity to stress intensity. In terms of a simple Paris power-law expression, da/dN $\propto$ ΔK^m, the exponent m was measured to be between 72 and 85. Increasing the temperature to 1100°C results in only a small increase in growth rates, with fatigue thresholds reduced from ~8 to 7 MPa√m in the coarse-grained material, and from ~6.5 to 6 MPa√m in the fine-grained material; the slopes of the growth-rate curves remain essentially the same (m ~ 79 to 82).

Compared to more traditional structural ceramics, such as Si_3N_4 (18-20), Al_2O_3 (21) and SiC (15), which have ΔK_{th} thresholds at 1000° to 1100°C of roughly 2 to 4 MPa√m (depending on material and testing conditions), the cyclic fatigue properties of Ti_3SiC_2 are clearly superior, both at low and high temperatures. However, at 1200°C, which is just above the ductile-brittle transition temperature of Ti_3SiC_2 (1), behavior is significantly different. Below ~10^{-8} m/cycle, growth rates are substantially faster, the slope of the growth-rate curves are reduced, and there is a sharp decrease, by a factor of ~ 2 to 3, in the fatigue thresholds in fine- and coarse-grained structures to ~2.3 and ~4.5 MPa√m, respectively.

Scanning electron microscopy images of the crack profiles during fatigue-crack growth in both structures, are shown in Figs. 3 and 4. What is apparent is that at 1100°C and below, i.e., below the ductile-brittle transition temperature, damage and shielding mechanisms are essentially unchanged. As shown for the coarse-grained microstructure in Fig. 3, the fracture mode at both 25° and 1100°C is predominantly intergranular, or "interlamellar" due to the delaminations within the grains (akin to a layered microstructure); in addition, approximately 10 to 20% of the fracture surface consists of transgranular/"translamellar" cracking. Similarly, the principal shielding mechanism in the crack wake remains grain bridging at both ambient and elevated temperatures (Fig. 3). Specifically, wake shielding can be observed both in the form of bridged grains and their frictional pullout and by an individual or clusters of lamella (typically deformed by bending) within the layer structured grains. This heavy deformation, together with the intrinsic strengths of the Ti-C bonds within the lamella (22), can increase the proportion of grains and lamellae participating in the bridging process, which acts to enlarge the extent of the bridging zone and hence increases the degree of toughening.

Typically, grain bridging and frictional pullout constitute the major source of extrinsic toughening (i.e., crack-tip shielding) in monolithic ceramics, such as Si_3N_4, Al_2O_3 and SiC (e.g., 9). In Ti_3SiC_2, the additional shielding process of the bending of deformed lamella appears to further enhance crack-growth

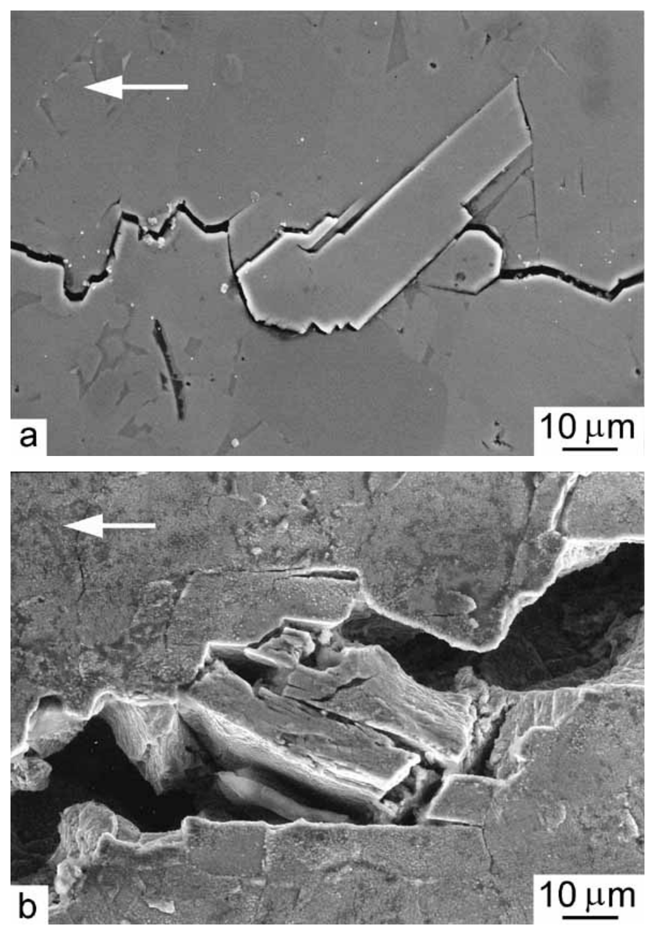

Figure 3: Scanning electron micrographs of the profiles of fatigue cracks propagating at (a) 25°C and (b) 1100°C in the coarse-grained Ti_3SiC_2 microstructure. Note the crack-wake grain bridging in both temperatures. Arrows indicate direction of crack growth.

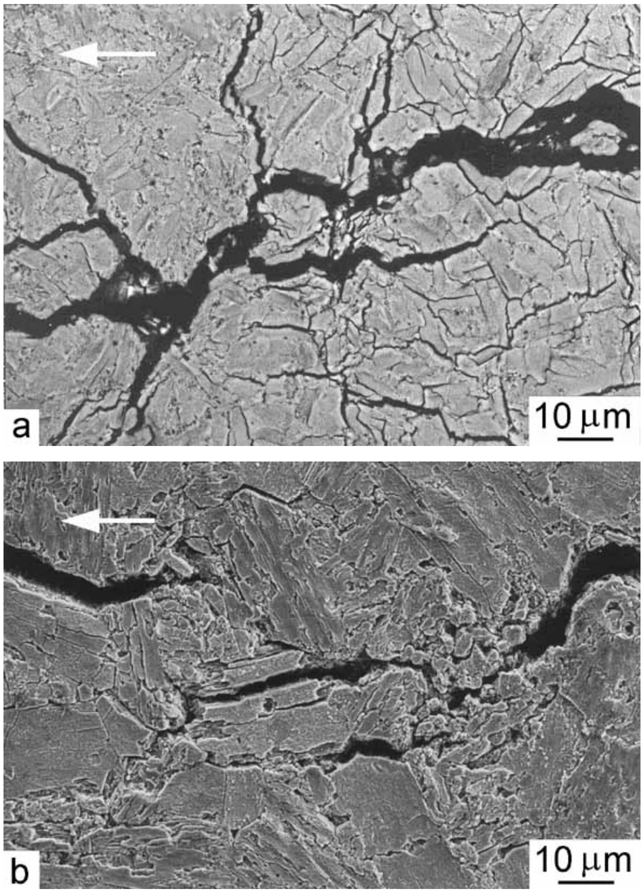

Figure 4: Scanning electron micrographs of the profiles of fatigue cracks propagating at 1200°C which is above the ductile-brittle transition temperature of Ti_3SiC_2, in (a) fine-grained, and (b) coarse-grained Ti_3SiC_2 microstructures. Note the profuse amounts of microcracks and cavities near the main crack. Arrows indicate direction of crack growth.

resistance, and hence the steady-state fracture toughness, by promoting very sizable bridging-zone lengths, which can exceed ~5 mm in the coarse-grained microstructure. Moreover, shear-faulting along the basal planes of the grain due to sliding along the contacting surface of a bridge may also increase crack-growth resistance by reducing the severity of frictional damage in the layered microstructures.

Below the ductile-brittle transition temperature, which has been estimated to lie between 1100° and 1200°C (1), no noticeable evidence of extensive damage, in the form of microcracking zones and cavitation ahead of the crack tip or viscous-ligament bridging by the grain-boundary glassy phase in the wake, could be seen. This strongly implies that the primary damage (intergranular/ lamellar cracking and the frictional wear degradation of the bridging zone in the crack wake) and crack-tip shielding (grain and lamellae bridging) mechanisms governing high-temperature fatigue-crack growth behavior in Ti_3SiC_2 up to ~1100°C are essentially unchanged from those at ambient temperature. The small decrease in crack-growth resistance at 1100°C (ΔK_{th} thresholds are ~8 to 14% higher at 25°C) may be rationalized by considering the nature of grain bridging (23) and its degradation under cyclic loading due to frictional wear (24,25). The pullout resistance from frictional tractions generated via sliding contact of opposing crack faces (26) is proportional to the normal stress acting on the interface, which in turn is a function of the residual stress resulting from thermal expansion anisotropy during cooling from the processing temperature. As the residual stresses will "anneal out" with increasing temperature, the normal stress will decrease, thereby reducing the pullout resistance.

Above the ductile-brittle transition temperature at 1200°C, the striking change in behavior, principally in the form of significantly reduced ΔK_{th} thresholds, can be attributed to the onset of significant high-temperature deformation and associated damage. There was noticeable macroscopic deformation of the test specimen (in the form of notch widening). More importantly, extensive microcracking and to a lesser extent cavitation was apparent throughout the sample, although the intensity of damage was most severe in the vicinity of the crack tip (Fig. 4). In addition, crack paths involved considerably more transgranular and/or translamellar cracking, especially in the fine-grained microstructure, compared to that seen at lower temperatures; this severely diminishes the propensity for grain bridging in the crack wake. It is worth noting here that since no new slip systems are activated above the ductile-brittle transition temperature, its atomistic nature is unclear at this time (1,11,22).

Thus, in summary, it can be seen that Ti_3SiC_2 represents an extremely damage-tolerant ceramic, with exceptional ambient temperature toughness and only a small degradation in fracture and fatigue properties at elevated temperatures below ~1100°C. Of the two structures examined, the coarse-grained microstructure displays a markedly higher K_c fracture toughness and ΔK_{th} threshold at both low and high temperatures, principally resulting from substantially more tortuous crack paths and extensive crack-wake bridging due to much larger bridging zones (typically ~ 4 – 5 mm in length in the coarse-grained structures compared to less than ~200 μm in the fine-grained structure).

Conclusions

Based on a study of cyclic fatigue-crack propagation behavior at elevated temperatures up to 1200°C in a reactively hot-pressed monolithic Ti_3SiC_2 ceramic with both fine- (3-10 μm) and coarse-grained (70-300 μm) microstructures, the following conclusions can be drawn:

1) The cyclic fatigue-crack growth properties of Ti_3SiC_2 were found to be superior to those of more traditional monolithic (non-transforming) structural ceramics, such as Si_3N_4, Al_2O_3 and SiC, both at ambient and elevated temperatures up to ~1100°C. Compared to behavior at ambient temperature, fatigue thresholds were only reduced by between ~8 and 13% at 1100°C; specifically, ΔK_{th} values were found to be ~6 and 7 $MPa\sqrt{m}$ in the fine- and coarse-grained microstructures, respectively. In both structures and at all temperatures below ~1100°C, growth rates displayed a marked sensitivity to stress intensity, with Paris power-law exponents varying from m ~ 72 to 85.
2) Mechanistically, damage and crack-tip shielding processes that were active at 1100°C were essentially unchanged from those at ambient temperature. Crack-path profiles showed a predominantly intergranular and/or interlamellar mode of crack advance with consequent crack-tip shielding by both grain and lamellae bridging in the crack wake. The lamellae bridging mechanism appeared to be particularly potent; as

this is not seen in more traditional ceramics, it may account for the superior damage tolerance shown by Ti_3SiC_2.

3) At 1200°C, above the ductile-brittle transition temperature, a striking change in the shape of the growth-rate curves was observed with ΔK_{th} thresholds being severely reduced (by a factor of ~ 2 to 3) to ~2.3 and 4.5 MPa√m, respectively, in the fine- and coarse-grained microstructures. Such behavior was attributed to the onset of significant high-temperature damage, in the form of macroscopic deformation of the sample, the presence of widespread microcracking and cavity formation, and the onset of some degree of transgranular and translamellar cracking which limited shielding by grain bridging in the crack wake.
4) Higher fracture toughness and cyclic fatigue-crack growth resistance were achieved in the coarse-grained Ti_3SiC_2 microstructure at both low and elevated temperatures. Indeed, this microstructure exhibited ~15 to 48% higher fatigue thresholds than the fine-grained microstructure at all temperatures between 25° and 1200°C. This is attributed to more extensive crack-wake bridging in the coarser structure, in the form of larger bridging zones and a greater tortuosity in crack paths.

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